

A semiclassical model for current-induced forces in electromigration and electroplasticity

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Abstract

We derive and employ a semiclassical spherical flow model to describe the atomic dynamics and calculate the current-induced force in the presence of electrical current. The conduction electrons serve as an ideal gas in classical mechanics flowing over atoms while using quantum mechanics for calculating the fluid pressure and viscosity. Two new kinds of current-induced force are derived based on present model, which are electron density force and electron viscous force. The calculated results are in good agreement with previous experimental results [Science 328, 736 (2010)]. Our model is also suitable for the motion and force analysis of nanoislands and voids. Combined with previous results, we firstly build the connection between electromigration and electroplasticity and help solve some long-standing puzzles.

Introduction.—Electroplastic effect was discovered by Troitskii and Likhtman¹. It was found that there was an immediate stress drop associated with application of a current during plastic deformation.^{1,2} The stress drop during electric current

application was termed the electro-plastic (EP) effect. This indicated that electric current might have a positive effect on material plasticity during plastic deformation, which had been broadly studied during the last two decades.³⁻¹⁹ In particular, there was an optimum strength of electric field which decreased the flow stress approximately 20%.⁴⁻⁵ and the mobility of metal defects (including dislocations, voids, grain boundaries) was found increased due to the applied large current.^{3,6,7,16-19} However, there were still some puzzles, such the mobility of dislocation increased both with or against the current direction during current application,¹⁶⁻¹⁹ which could not be well explained. On the microscopic scale, the large current in a metal can generate preferential diffusion of atoms in one direction. This phenomenon was called electromigration.^{20,21} The effect of current-induced forces in atomic and molecular wires has been studied in recent work.²²⁻²⁷ Though the current-induced force had been broadly measured or calculated, however, there's still a huge difference between the predicted value and the experimental results due to the complexity of many body problems. Also, some phenomenon cannot be well explained, such as the grain boundary drag effect,²⁸ the motion of nanoislands.²⁹ A connection between them is also fail to be built due to topological problem (dislocation) and the complexity of atomic structure.

A unified model including all aforementioned effects is highly desirable for further exploration in this field. In this article, the conduction electrons are degenerate, free, non-interacting particles, which serve as an ideal gas in classical mechanics, with the pressure and viscosity determined by quantum mechanical effects. Two new kinds

of current-induced force are derived and they can be roughly represented as $3\rho_e haV$ and $\Delta\rho_e K$. Despite its seemingly simplicity, the calculated results agrees surprisingly well with previous experimental results, which is even much more accurate than DFT calculation results. These two kinds of force can also be applied to different models to explain some puzzles. For example, combined with Frenkel-Kontorova model, the connection between electromigration and electroplasticity is firstly built. The dislocations are driven by the force that is exerted on atoms, which is completely different from conventional deformation mechanisms and help explain some puzzles.

Electron density force (EDF).—The atom movement in the electron gas is reduced to the classical movement of a sphere in unbounded ideal fluid. We mainly analyze the movement of atoms in dislocation line. The atom in the dislocation line (hereafter called dislocation atom) is the sphere, and the electron gas is the unbounded ideal fluid. The fluid is the relative reference frame, and the sphere moves in the fluid with velocity V ($V = v_{ato} - v_e$) , where v_{ato} is the atom velocity, v_e is the free electron drift velocity. On the sphere surface, the distribution of relative velocity is

$$v_{r=a} = \frac{3}{2}V \sin \theta , \text{ where } \theta \text{ is the angle between the radius and x-coordinate.}^{30}$$

Average the horizontal component of velocity of the left side of sphere, we arrive at

$$\bar{v} = \frac{1}{\pi} \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{3}{2}V \sin^2 \theta d\theta = \frac{3}{4}V \quad (1)$$

Similarly, the velocity of the other side can be got as $\frac{3}{4}V$. Assuming that A is at the left side of dislocation and B is at infinity. The mean horizontal velocity of the electron gas at A is v_A , the horizontal velocity of the electron gas at B is v_e , which is

the electron gas drift velocity. The pressure of the electron gas at A is P_A . The pressure of the electron gas at B is $P_B = P$ and $\rho_B = \rho$, where P is the mean electron gas pressure, ρ is the mean electron gas mass density. Each side of the dislocation atom is assumed to meet Bernoulli equation with different density, respectively. The pressure equation for electron gas at A is obtained as:

$$P_A = \frac{\rho_A}{\rho} P + \rho_A \frac{v_e^2 - v_A^2}{2} \quad (2)$$

Similarly, for the other side C

$$P_C = \frac{\rho_C}{\rho} P + \rho_C \frac{v_e^2 - v_C^2}{2} \quad (3)$$

where $v_C = v_A = \frac{3}{4}V + v_{ato} = \frac{3}{4}v_e + \frac{1}{4}v_{ato}$. According to D'Alembert paradox, the pressure of both sides should be the same. However, the electron gradient between two sides of the atoms in the dislocations line leads to the electron density force. The direction of EDF is only determined by the electron density gradient. Then, the expression for EDF can be obtained as

$$\sigma_{EDF} = P_A - P_C = \frac{\Delta\rho}{\rho} P + \Delta\rho \frac{v_e^2 - \left(\frac{3}{4}v_e + \frac{1}{4}v_{ato}\right)^2}{2} \quad (4)$$

where $\rho = \rho_e \cdot m_e$, ρ_e is the electron distribution density, m_e is the electron mass. Then the basic equation for EDF is obtained as follow

$$\sigma_{EDF} = \frac{\Delta\rho_e}{\rho_e} P + \Delta\rho_e m_e \frac{v_e^2 - \left(\frac{3}{4}v_e + \frac{1}{4}v_{ato}\right)^2}{2} \quad (5)$$

For conduction electrons, only those very close to the Fermi energy can participate in the conduction process. Therefore, their pressure can be got by small

deduction as $P = \frac{2}{5} \rho_e E_f$ where E_f is Fermi energy level. For the second part of equation (5), it derives from the kinetic energy from electrons to ions. It can be replaced by

$$\rho_e v_e \sigma_{sca} \tau_{col} E_f = \rho_e v_e \sigma_{sca} \frac{l_{mfp}}{v_e} E_f = E_f \quad (6)$$

where σ_{sca} is the scattering cross-section of ion, τ_{col} is the mean time interval between two successive collisions, $l_{mfp} = (\rho_e \sigma_{sca})^{-1} \cong \rho_e^{-1/3}$ is the mean free path between electrons and ions. Assume the atom velocity in current direction is zero.

Then equation (5) can be rewritten as

$$\sigma_{EDF} = \frac{2}{5} \Delta \rho_e E_f + \frac{7}{16} \Delta \rho_e E_f \cong 0.84 \Delta \rho_e E_f \quad (7)$$

$\Delta \rho_e$ is the electron distribution density gradient between two sides of dislocations. Electron density force is like the nature wind which is created by the air density difference/gradient. The distribution of electron is significantly affected by various factors such as charged vacancies and congregated solute atoms.³¹⁻³⁵ Between the two sides of dislocation, under the effects of charged vacancies, a vivid contrast of electron distribution density is formed. The electron density gradient has been observed in previous researches. The constrictions in current flow path can increase local density up to about a factor of 2.³⁶ Also, atomic relaxation at step edges redistributed local charge density,^{37,38} with enhancement up to a factor of 10 on Cu(532) near kink sites.³⁹

An equation for roughly estimating dislocation charge polarization is proposed based on the assumption that gliding dislocations sweep up charged vacancies.⁴⁰ For a dislocation pileup of $2l_0$ length locked at $x = \pm l_0$, the charge distribution density can

be written as follows

$$\rho_{char}(x,t) = A_0 \exp(-rt^2) \left\{ \frac{2}{x^2 + \alpha^2(t)} - \frac{1}{(x+l_0)^2 + \alpha^2(t)} - \frac{1}{(x-l_0)^2 + \alpha^2(t)} \right\} \quad (8)$$

where A_0 is the amplitude of the vacancy charge distribution at initial moment ($t=0$); r is supposed to be a constant. The dislocation is at initial moment $t=0$, the density equation is thus

$$\rho_{char}(x,t) = A_0 \left\{ \frac{2}{x^2 + \alpha_0^2} - \frac{1}{(x+l_0)^2 + \alpha_0^2} - \frac{1}{(x-l_0)^2 + \alpha_0^2} \right\} \quad (9)$$

The dislocation pileup consists n dislocations, and the dislocation is equidistant from each other. Then, the x coordinates of the top dislocation and the next dislocations are $x_1 = l_0$ and $x_2 = l_0 - 2l_0/n$, respectively. The charge density gradient $\Delta\rho_{char}$, which is proportional to $\Delta\rho_e$, can be obtained as

$$\Delta\rho_e \propto \Delta\rho_{char} = \rho(l_0, 0) - \rho(l_0 - 2l_0/n, 0) \quad (10)$$

Equation (10) builds a connection between plastic deformation and electron density gradient. During plastic deformation, the electron density gradient is mainly determined by charged vacancies or solute atoms which are swept up by moving dislocations. The electric field or current can change direction of electron density gradient, thus forming new motive force for atom or dislocation motion. This prediction is recently demonstrated by reference 41. Using DFT calculation, the charge density gradient of Ag can be represented as:

$$\int \delta\rho_e dx = \int (\mu_L - \mu_R) [\rho_L(\mu_{eq}, r) - \rho_R(\mu_{eq}, r)] dx / 2 \quad (11)$$

And the simulation results indicates that could be of $4 \times 10^{-27} \text{ m}^{-3}$ on chain and dislocation lines, which is directly suitable for present model.

The current-induced stress is about a few GPa.^{7-9,13} In reference 41, $\Delta\rho_e$ is firstly calculated using first principles recently and $\Delta\rho_e$ could be of $4\times10^{27} m^{-3}$. The conduction electron density ρ_e for single-crystal Ag is $5.85\times10^{28} m^{-3}$, Fermi energy is 5.48eV, therefore, according to equation (7), $\sigma_{EDF} = 2.94 Gpa$. Equation (7) contains two parts. The left part derives from the electron density gradient, while the other part derives from the quantum kinetic energy transfer. We use classical fluid mechanics to deduce the basic equations for EDF. Then, we further transform it to quantum mechanical forms, and the calculated results demonstrate the success for the transformation we have made.

Electron viscous force (EVF).—The other kind of current-induced force is caused by electron viscosity. In classical fluid mechanics, the viscous drag formula has been given by Navier-Stokes³⁰

$$F = 6\pi\mu aV \quad (12)$$

where V is the relative velocity of the sphere to the fluid and $V = v_{ato} - v_e$, μ is dynamic viscosity, a is the sphere radius. We assume the conduction electrons as degenerate matter (Fermi gas). A Fermi gas of weakly coupled systems obey universal hydrodynamics.^{42,43} The viscosity has a universal quantum scale $\rho_e \hbar$, where \hbar is Planck's constant h divided by 2π . A more accurate form of this viscosity can be got as

$$\mu = \alpha_\mu \rho_e \hbar \quad (13)$$

where α_μ is a universal dimensionless coefficient, the electron viscous force can thus be rewritten as

$$3\alpha_\mu \rho_e h a V \quad (14)$$

It is worth noting that equation (13) is applicable to conduction electrons when the mean free path l_{emfp} of electrons is much smaller than the characteristic size R of the system. The so-called Knudsen number is $K = l_{emfp} / R$.⁴² For conduction electrons, the characteristic size is determined by the de Broglie wavelength $\lambda = h / \sqrt{2m_e E_f}$ and $l_{emfp} \cong \rho_e^{-1/3}$. $l_{emfp} / \lambda = 0.49$.

We then choose the value in reference²⁹ to make a comparison between the predicted EVF value and the measured one. According to $v_e = \frac{j}{\rho_e e}$, when j is $6.7 \times 10^9 \text{ A/m}^2$, v_e is 0.715 m/s . The ratio of mean energy per conduction electron to Fermi energy E / E_f is about 1, according to reference⁴², the viscosity coefficient α_μ is $2.30 \sim 2.50$, then, the dynamic viscosity is $\mu = (1.42 \sim 1.54) \times 10^{-5} \text{ pa}\cdot\text{s}$. The atom radius of Ag^+ is $1.26 \times 10^{-10} \text{ m}$, then $F_{EVF} / j = (2.25 \sim 2.44) \times 10^{-3} \text{ eVcm/A}$. Compare with the measured value $F / j = (2.2 \pm 0.9) \times 10^{-3} \text{ eVcm/A}$.²⁹ However, the expected values based on previous models using first principles are $3.0 \times 10^{-5} \text{ eVcm/A}$ and $6.6 \times 10^{-5} \text{ eVcm/A}$,^{35,36} and there's still lack of an accurate and effective method for calculating the current-induced force at present²⁹. We introduce fermions viscosity into the classical stokes drag formula and it's surprising that the calculated value is of such high accuracy.

Chenggang Tao et al visualized the electron scattering force in Nanostructure by observing the current-biased displacement of monatomic islands of several nanometers on single-crystal Ag.²⁹ It was found that the velocity of the islands varies inversely with radius, which is contradictory to the prediction of pure

attachment/detachment mechanism. However, this phenomenon can be well explained by our theorem. Assuming the nanoisland is a disk, then the drag force of the disk is $F_{disk} = -\frac{32}{3} \mu V R_0$,³⁰ the island area density is ρ_{is} , the island radius is R_0 , the island drift velocity is v_0 .

$$\frac{dv_0}{dt} = \frac{32\rho_e \hbar (v_e - v_0)}{3\rho_{is}\pi R_0} \quad (15)$$

Then, we can get

$$v_e - A e^{-\frac{32\rho_e \hbar}{3\rho_{is}\pi R_0} t} = v_0 \quad (16)$$

Because $v_0 = 0$ when $t = 0$, then $A = v_e$, therefore

$$v_0 = v_e (1 - e^{-\frac{32\rho_e \hbar}{3\rho_{is}\pi R_0} t}) \quad (17)$$

$$\frac{\partial v_0}{\partial R_0} = -\frac{32\rho_e \hbar v_e}{3\rho_{is}\pi R_0^2} e^{-\frac{32\rho_e \hbar}{3\rho_{is}\pi R_0} t} \quad (18)$$

According to equation (18), the island drift velocity v_0 varies inversely with radius R_0 , which well suits the experimental results. Additionally, according to equation (18), the island drift velocity will never be larger than the electron drift velocity. This phenomenon is thus explained in a completely different way from the diffusion mechanism. An important observation was made by K.C. Chen et al.²⁸ They found that the grain boundary slowed down electromigration rate by one order of magnitude. According to equation (11) and (7), this time the electromigration direction might be opposite to the electron density gradient in grain boundary, which leads to the drag effect.

Application.—While previous investigation had proposed the concept of ‘electron

wind force' which might help promote the deformation,^{4,10} they failed to demonstrate that electric current could exert force on topological defects (dislocation). In Frenkel-Kontorova model, the dislocation motion which is driven by dc (direct current) force can be represented as⁴⁴

$$\ddot{u}_l(t) + \eta \dot{u}_l(t) + \sin u_l - g(u_{l+1} + u_{l-1} - 2u_l) = F \quad (19)$$

where the dot stands for the time derivative, η is the damping coefficient, u_l is the coordinate of the $l-th$ atom, g is the elastic constant, F is the applied dc force to all atoms. The dc-driven FK model was investigated by a series of papers by Braun et al.⁴⁵⁻⁴⁷ A similar ac-driven dislocation motion has also been given by previous researches.⁴⁸⁻⁴⁹ However, while the FK model demonstrate the effect of dc current on dislocations, it is lack of an effective representation of dc force. Combined with our model, dc force F in equation (19) can be replaced by equation (14) as

$$\ddot{u}_l(t) + \eta \dot{u}_l(t) + \sin u_l - g(u_{l+1} + u_{l-1} - 2u_l) = 3\alpha_\mu \rho_e h a (V_e - \dot{u}_l(t)) \quad (20)$$

according to equation (14), the plastic deformation is promoted by EVF. If the drive force is EDF, according to equation (7) and (11), EDF can help explain the increased mobility of grain boundary.⁷ It had been pointed out that current-driven motion of the dislocation is possible if the drive's amplitude exceeds a threshold value.^{48,49} This, combined with equation (20), can help explain why there is an optimal strength of electric current⁴⁻⁵ and can help define the threshold value of current density. The electron wind force-driven dislocation motion had been directly observed and there existed an threshold value of the applied electric field for driving these dislocations.⁵⁰ According to equation (20), EVF and EDF provide extra motive

force for dislocation motion. They thus help depin dislocations and reduce double-pinned Frank-Read sources, thereby reducing dislocation density. This explains the dislocation density reduction effect.^{14,15}

In reference 51,52, the solution has been obtained for the motion of void driven by electric potential and gradient stress field. If we consider the effect of EDF, which results from the electron density gradient of the void, the chemical potential of the atoms on the interface should be

$$\mu = \mu_0 - \Omega \gamma_s \kappa - Z^* e U - \Omega \sigma_{EDF} - \Omega \sigma_n + \Omega U_s \quad (21)$$

where μ_0 is the reference value of the potential, γ_s is the interface energy, κ is the curvature of the interface, Ω is the atomic volume, U is the applied electric potential, U_s is the strain energy density of matrix material, σ_n is the normal stress acting on the interface. The atomic flux along the interface is given by

$$J = -\frac{D_i \delta_i}{\Omega kT} \frac{\partial \mu}{\partial s} = \frac{D_i \delta_i}{KT} \left(\gamma_s \frac{\partial \kappa}{\partial s} - Z^* e E_i + \frac{\partial \sigma_{EDF}}{\partial s} + \frac{\partial \sigma_n}{\partial s} - \frac{\partial U}{\partial s} \right) \quad (22)$$

where D_i is the interface diffusivity, δ_i is the thickness of the interface, kT has the usual meaning, s is the arc length of the interface, $E_i = -\nabla_s U$. The mass conservation requires that $dJ/dy = V/\Omega$, then integrate it from y to $y = d\sqrt{(1-m)/(1+m)}$, where m is the shape factor, we get

$$J = \frac{V}{\Omega} \left(d \sqrt{\frac{1-m}{1+m}} - y \right) + C \quad (23)$$

where the constant C can be determined by boundary condition, m is the shape factor, then

$$C = -\frac{D_i \delta_i}{kT} \left(Z^* e E_i + \nabla_s \sigma_{EDF} + \frac{\partial \sigma_n}{\partial s} - \frac{\partial U}{\partial s} \right) \quad (24)$$

where $\frac{\partial \sigma_{EDF}}{\partial s}$ depends on electron density gradient. Under the effect of gradient electron density gradient, atoms diffuse along the interface from one side of high density to another of lower one. Therefore, $\frac{\partial \sigma_{EDF}}{\partial s} = -\nabla_s \sigma_{EDF} < 0$. Symmetry requires that the flux at $y=0$ must be zero, thus

$$V = \frac{\Omega D_i \delta_i}{dkT} \sqrt{\frac{1+m}{1-m}} \left(eZ^* E_i + \nabla_s \sigma_{EDF} - \frac{\partial \sigma_n}{\partial s} + \frac{\partial U}{\partial s} \right) \quad (25)$$

According to equation (25), electron density force contributes to the motion of voids.^{3,6} Additionally, EDF helps explain the variation of the void velocity which might result from the variation of electron density distribution.^{3,6} It had been shown that dislocations increase their velocities when moving both with and against the current and the dislocations move along current direction approximately 10-20% faster than against it, which is called non-polarization effect.¹⁶⁻¹⁹ The improvement of dislocation mobility is the result of various effects including EDF and EVF and the other part in equation (25), such as the gradient stress field.^{51,52} the plasticity can be improved even when EVF is adverse.

Conclusion.—The current-induced force had been called “electron wind force” for a long time, however, it was merely a concept. Now, in present model, this force is finally demonstrated as a real electron wind. Our model assumes the electron gas as Newton fluid, which is a very bold assumption and completely different from previous researches. However, the calculated results and well explanation of relative phenomenon strongly corroborates our assumption. We calculate the electron force using Fermi viscosity, Fermi pressure, Navier-Stokes formula and Bernoulli formula, and the calculated value agrees well with experimental value. We have also firstly

built the connection between electromigration and electroplasticity combined with previous results, by which some long-standing puzzles are solved. Our model is also suitable for the motion and force analysis of materials of various scales, such as atoms, nanoislands, dislocations and voids.

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